We thank the reviewers for their helpful comments on our manuscript. Below, please find our responses in blue and the reviewers' comments in black. The line numbers in our comments refer to the line numbers in the revised manuscript.

Reviewer 1:

In this manuscript the authors have assembled a high quality dataset of smoke observations and evaluated the ozone production from these with both a global and box model. The authors work has led to several insights into smoke chemistry, and as such, will be a useful addition to the literature on this topic. However I have a number of comments on the methodology and suggestions on the overall presentation.

We thank the reviewer for their comments and suggestions.

Major points:

1. In section 2.4 they describe scaling of photolysis rates to examine the impact of aerosols on photochemistry. It seems they use an equal scaling for all photolysis rates. This is probably not right as some are more sensitive to aerosol scattering and absorption then others, depending on the wavelength. Can the authors comment on this or describe how this would impact their analysis.

Thank you for bringing this up for discussion – a more detailed treatment would require accounting for wavelength-dependent aerosol extinction, which is beyond the scope of this study. Our use of a single uniform scaling factor was intended as a simple test of how reduced actinic flux from smoke aerosol could alter plume chemistry, rather than a physically exact representation of aerosol radiation interaction (as one might perform in a 3D model). The goal of the box modeling work is a sensitivity comparison between different factors impacting ozone formation in smoke plumes.

We now clarify this assumption in the supplementary text (Box model description) and have added a short discussion of how wavelength-dependent aerosol impacts on photolysis could alter specific reactions:

> "By ignoring the wavelength-dependence of aerosol extinction in our approach, we only sample a portion of the potential photolysis distribution, but we argue that this constitutes an approximate mean photolysis suppression behavior. We believe that the key result of the box modeling analysis, that the aerosol optical effect is more important in governing ozone production than the chemical effect, would not be impacted by a more thorough treatment of wavelength-dependence."

2. Similarly, for the idealized box model, it seems the SZA is fixed at zero. Can the authors discuss the impacts of this assumption.

As stated above, we intend this section as an idealized study of plume chemistry and the relative importance of different chemical factors in governing ozone production. By fixing the SZA at 0°, we are analyzing the chemistry in a hypothetical plume observed around noon.

To examine the impacts of this assumption, we re-ran our set of simulations at a different SZA (45°). The results of this sensitivity test are shown in Figure S10. While the magnitude of PO3 changes slightly with the SZA change, the relative spread associated with each factor remains similar in both cases. See line 230:

> "Default photolysis rates are kept constant and calculated using the simplified MCM parameterization assuming a solar zenith angle of 0°, simulating clear-sky photolysis with the sun directly overhead. Temperature is held constant at 288.15 K. Our findings are robust to variations in temperature and solar zenith angle, as shown in Fig. S10."

3. In Figure 3, the authors an enhancement in NOx/CO at high photochemical age. This is a bit surprising. Is this due to PAN decomposition? How would this plot look if NOy were used instead of NOx. What is the role of PAN decomposition on ozone production?

We believe the reviewer may have mis-interpreted Figure 3. In this figure, we show production of ozone as a function of photochemical age (not NOx/CO as a function of photochemical age). We split the data into two segments using the OH reactivity weighted ratio between NOx and CO, θ' "to approximate ozone production sensitivity to its precursors" (L277). There is no enhancement in NOx/CO at high photochemical age, but there is enhanced ozone production in points with higher NOx/CO (θ'). We hope that by introducing the θ' parameter earlier, in Section 2.6, this distinction becomes clearer.

4. The discussion on photolysis is mostly good. But I am left with a question of whether this photolysis suppression impacts the overall ozone production? Perhaps this photolysis suppression of ozone is a short term effect, which simply prolongs the NOx lifetime and therefore has little (or less) impact on the overall ozone production. Comments?

This is an interesting point. We are limited by the instantaneous nature of the measurements. This could perhaps be explored in a case study (plume-by-plume) approach. We have now added text to this effect on Line 383.

> "From our analysis, we cannot conclude how much local ozone production suppression reduces the overall O_3 production in the lifecycle of a fire plume; future plume-following case study analysis may be able to further explore this question."

5. In the abstract and elsewhere, the authors describe several key limitations of Geos Chem; too high ozone production and too little biomass burning emissions. It is not clear if these "two wrongs make a right". In other words, can we assume that GC is getting the right answer due to offsetting errors? Can the authors describe more quantitatively how well these errors offset?

Indeed, errors in GEOS-Chem do appear to fortuitously cancel to some degree (as stated on Line 496). We cannot be more quantitative about this conclusion given that neither a “correct” emissions inventory (in terms of magnitude and variability), nor “correct” chemical production mechanism in GEOS-Chem are available to us. Thus, we cannot quantify the relative role of errors in emissions and errors in chemical production at this time.

Other points:

Line 35 (abstract): Is this reduction in ozone production temporary or permanent?

Thank you for pointing out this ambiguity. The text has been clarified:

> “Observations in fresh smoke demonstrate that suppressed photochemistry reduces O_3 production by ~70% in units of ppb O_x (O_3+NO_2) per ppm CO in the near-field (age<20hr).”

L37: Sentence beginning with “Using a box model…” This needs rewriting. Hard to follow this very long sentence.

Thank you for your suggestion. We have split the sentence into two sentences in the new version (L37):

> “Using a box model, we explore the evolving sensitivity of O_3 production to fire emissions and chemical parameters. We demonstrate the importance of aerosol-induced photochemical suppression over heterogeneous HO_2 uptake, validate HONO’s importance as an oxidant precursor, and confirm evolving NO_x sensitivity.”

L42: Sentence starting with “these discrepancies” describes the problems, but then next sentence ignores the problems. We need to better understand if we can believe the GC results here.

In this work we expose some limitations of the model (which have not been previously identified), but show that the model does reasonably reproduce observed ozone enhancement levels. We change L42:

> “These discrepancies drive biases in normalized ozone production ($\Delta O_3/\Delta CO$) across plume lifetime, though the model generally captures observed absolute O_3 enhancements in fire plumes.”

We therefore show what the model estimates as the ozone contribution from fires world-wide. We believe that we have fairly documented the limitations, which may be more important when attempting to use the model to mechanistically explore ozone production. After the evaluation of potential uncertainties that we provide in Section 3.3.2, the reader can make an informed interpretation of the GEOS-Chem estimate. This is highlighted in the first sentence of Section 3.3.2 (L541):

> "In Figure 9, we quantify the overall impact that fires have in governing the tropospheric ozone burden in the GEOS-Chem model."

And, we have added additional text to address some of these concerns (L556):

> "It is worth noting that fire emissions exhibit interannual variability (Cooper et al., 2024) and that fire emissions inventories struggle with regionally-dependent uncertainties (Wiedinmyer et al., 2023). The reported results describe GEOS-Chem's representation of fire impacts on global ozone in a single year (2020), subject to the uncertainties in fire emission inventories."

L110: Note that Lee and Jaffe extended this analysis to include more than 600 sites in the continental US.
https://doi.org/10.1021/acs.est.4c05870

Updated citation added, thank you. L114 now reads:

> "Lee and Jaffe (2024) employ a statistical approach to estimate that wildfires contribute ~7.8 ppb of additional surface ozone on smoky days compared to no-smoke days at surface measurement sites across the U.S."

L154: If I am understanding the units here, I am not sure this excludes all stratospheric airmasses. For example, an airmass with O_3 = 500 ppb and water vapor of 2 g/kg would have a delO3/H2O ratio of around 0.4 ppb/ppm and so would be included. Comments?

You are correct that such outliers would theoretically slip through this filter (taken from Bourgeois et al. 2021 for consistency with their analysis). However, Figure S1 shows that no observations with ozone above 150 ppb is included in our final dataset.

L195: Using the emission factors to get the t=0 ratios seems like a big assumption, given large plume to plume variability. What is the uncertainty in this? Why can't you use the observations directly to get the t=0 values?

Previous work, such as Gkatzelis et al. 2023, Permar et al. 2021, and Fiddler et al. 2024, have attempted to estimate emission factors directly from airborne observations, as suggested by the reviewer. They do find that plume to plume variability is large, and is shown to be related to burning conditions, fuel type, diurnal variability, and more. However, all these estimates tend to agree within reasonable uncertainty. Thus, while assuming a consistent emission factor for t=0 is imperfect, we think it is a reasonable assumption when integrating a large dataset of fires. Please see the discussion on uncertainty at L214.

> "The accuracy of the photochemical age estimate is limited by (1) the assumption of constant [OH] during a plume's lifetime, (2) the uncertainty in VOC emissions ratios and

> variation arising from diverse fuel types, different burning conditions, and plume-to-plume variability (3) the measurement uncertainty in our VOC observations, and (4) potential mixing between more and less aged air masses. Given this, we use the derived photochemical ages to qualitatively measure the evolution of a smoke plume, and the results of our study depend on relative rather than absolute ages, as discussed below."

L210: There are a number of simplifying assumptions here that need to be discussed as to implications. Mainly fixed SZA at 0 degrees and scaling all photolysis frequencies the same, despite some mainly in the visible part of the spectrum, while others are more related to UV.

We thank the reviewer for highlighting these points above – please see our previous responses (these assumptions are now discussed in SI).

Figure 3: It took a few minutes to figure this figure out. It would help to discuss phi in the context of high and low NOx regimes. There are some points at very high O_3/CO ratios (light blue). I wonder if these might be better removed with a lower delO3/H2O screening ratio? As noted above, it is surprising to see high NOx values at large photochemical ages (eg 50-200 hours). Is this related to PAN decomposition? Can you show this plot with NOy instead of NOx?

We thank the reviewer for highlighting these points above – please see our previous response.

L295: These equations are out of place. They should be moved to before Figure 3. Also, as noted above, the phi values are approximately equal to k*Conc(NOx)/Conc(CO). Thinking about these as concentration ratios makes it a bit easier to conceptualize what is happening here…

We have moved the equations and associated text to Section 2.6 in Materials and Methods. Theta prime is a ratio of rates (and thus an ozone production regime metric), so should not be interpreted as a ratio of concentrations.

L318: Suggest to "observed J-NO2". As noted above, need some discussion on whether this is a temporary or permanent reduction in O_3 production.

We thank the reviewer for highlighting this point above – please see our response there.

Figure 5: It is interesting that you see enhanced NOx in the fire regimes. This contrasts with the work of Buysse 2019 (DOI: 10.1021/acs.est.9b05241). Can you discuss in more detail?

The fire regime is defined using long-lived organic tracers (CH3CN, HCN) in an effort to include both fresh and aged smoke. Within our dataset most of the fire regime is observations of fresh smoke taken from the WE-CAN and FIREX-AQ campaigns, as addressed in L352:

> "In contrast, the majority of our fire influenced observations are taken in the near-field (67%) and are rich in NO_x, as reflected in Fig. S9."

It appears that the dataset compiled in Buysse et al. consists of EPA AQS data from 18 western U.S. cities combined with NOAA HMS data to attribute smoke influence. We would hypothesize that the smoke observed at these ground sites in urban environments is much older that the near-field smoke sampled in the airborne dataset we compile. We believe this is the reason that the mean NOx behavior differs between the two studies.

Figure 6: Suggest a statistical comparison of O_3 concentrations for the points with higher TCE values…

Thank you for your suggestion. A comparison of O_3 concentrations and other NOy species is shown in Figure S12.

L370: This whole paragraph is confusing and needs rewriting.

We are sorry that the reviewer found this paragraph confusing. We have re-visited the text and without more specific guidance from the reviewer, we cannot identify what they found confusing.

L390: Didn't you also test something having to do with NOx?

One of the factors we tested was the NOx/CO ratio. We find that it increases in importance with age, matching the observed results in Figure 3. We add this phrase to L433:

> "We use our box model to test the sensitivity of ozone production against specific factors including (1) the speciation and reactivity of the emitted *VOC* population, (2) the range of wildfire emissions observed across fires that vary in size, fuel, and burning conditions, (3) the emission of radical precursors, (4) the evolving availability of NO_x, and (5) aerosol effects."

Figure 7 is hard to decipher. A lot going on here. Suggest to simplify to emphasize the main points.

Figure 7 does contain quite a bit of information. Panel c) shows the key result, however we feel that moving panels a) and b) to the SI would force the reader to open this extra document to interpret the results. Thus, we prefer to keep all the panels together.

L423/section 3.3:
So this is where we need some discussion on the GC discrepancies and how the two major issues (ozone production and too little bb emissions) balance.

We thank the reviewer for highlighting these points above – please see our previous responses.

L465: I am not seeing this in the Bourgeois analysis. Can you clarify?

Bourgeois focused on far-field, aged smoke observed in ATom. They found that modeled wildfire-O3 was underestimated. See the fourth paragraph of the results section titled "Comparison of Observed and Modeled BB Influence on Tropospheric O3", which begins:

> "None of the models adequately represents the strong influence of BB emissions on tropospheric O3…"

This result is reflected in Figure 5, which shows that ATom observations ascribe more ozone to biomass burning influence relative to urban influence, while GEOS-Chem and other models do the opposite.

Figure 9: This is good representation of the results, but we need more details. What years does this represent, what months/ozone season and what are the area boundaries?

Thank you for the comment. We have added information to the Figure 9 caption:

> "Figure 9. Impact of wildfire emissions on tropospheric and surface ozone in GEOS-Chem for the year 2020. (a) Percent change in the tropospheric ozone column burden due to wildfire emissions, with the largest enhancements over tropical Africa and South America. (b) Percent of surface ozone concentrations attributable to wildfire emissions. (c) Regional, population weighted surface ozone exposure. The regions in (c) correspond to the colored / patterned outlines in (b)."

L530: Please add "and under-estimates biomass burning emissions " or something along this line.

Thank you for the suggestions. This point is addressed in L576: "In the near-field, we show that GEOS-Chem cannot capture concentrated fire plumes, and that the model cannot capture the NOx-suppressed and photochemically suppressed sub-grid conditions." It is difficult to say for sure that emissions are underestimated, because resolution also plays a role in diluting emissions in each grid cell.

Reviewer 2:

GENERAL
This study provides valuable insights into wildfire-driven tropospheric ozone production by integrating airborne observations, box modeling, and global-scale simulations with GEOS-Chem. While the work makes a meaningful contribution to understanding of fire plume chemistry and its impacts on atmospheric ozone, several limitations and uncertainties remain that warrant further consideration.

We thank the reviewer for their comments and suggestions.

First, the reliance on airborne measurements, though offering unique perspectives, is inherently constrained by the limited temporal and spatial coverage of individual campaigns. Wildfires are highly heterogeneous in terms of fuel type, combustion phase, meteorological conditions, and plume dynamics, all of which can strongly influence ozone chemistry. The study synthesizes results from several campaigns, but it is unclear whether the dataset fully represents the diversity of global fire regimes, especially those occurring in tropical regions and the Southern Hemisphere, which are under-sampled compared to North American events (see Fig. 2).

Thank you for your comments. The campaign data that we have access to is indeed more North American centric. We add some discussion to address this point (L570):

> "More observations are needed particularly in regions outside the US – the campaign data that we synthesize has poor coverage of the Southern Hemisphere and does not fully represent the fuel types and meteorological conditions outside North America."

In addition, the airborne sampling indicates ozone formation process at the height of several kilometers, missing the information near the surface. This raises questions about the broader generalizability of the conclusions.

By using a background subtraction and using photochemical clocks to put our dataset in terms of smoke age, we hope to extract conclusions that can be generalized across different plumes at varying heights. As your comment supposes, the chemical behavior of smoke plumes does indeed change significantly during the aging process, as we discuss in detail. In our opinion, the main takeaways of the study are: 1) the chemical regime transition from NO_x-saturated to NO_x-limited that occurs during smoke aging, 2) the importance of suppressed photochemistry in limiting ozone production under heavy smoke in the near-field, and 3) mixed fire-anthropogenic air masses display higher ozone production than air masses that are solely fire-influenced. While it is true that airborne sampling cannot capture plumes at all altitudes and ages, we believe that these conclusions are generally applicable to every smoke plume.

For example, if your comment about altitude is related to temperature – while ozone chemistry is temperature dependent as temperature will change the rate of certain reactions and the rate of ozone formation, the relative importance of different chemical factors that we discuss: NOx availability, oxidant availability, aerosol effects, etc., is a conclusion that can be generalized across plumes of different heights. This is discussed in more detail in response to one of your comments below, and in Figure S10.

Second, the evaluation of GEOS-Chem highlights important discrepancies, such as the model's overestimation of near-field ozone enhancements and inability to capture the chemical regime transition. However, the analysis seems largely diagnostic rather than mechanistic, as it does not indicate whether targeted sensitivity tests within GEOS-Chem were performed to isolate specific causes (e.g., emission inventories, plume injection heights, or chemical mechanisms).

Indeed, we have only evaluated the GEOS-Chem performance. Having used this unique dataset to explore ozone production in fire-influenced air masses, we felt it was an opportunity to evaluate a 3D model representation of the same. We did not perform targeted sensitivity tests, though they would be very interesting to pursue in future work. We believe that our text accurately reflects that we did not use GEOS-Chem to interpret the observations, but rather that we used the observations to test the model, for example our abstract states "We evaluate GEOS-Chem's performance against these observations…"

In addition, the coarse spatial resolution (2x2.5 or 0.5x0.625) can not resolve the ozone changes in the near-field plumes. Even though the model captures the ozone enhancement in the aged or far-field plumes, the derived sensitivity may not warrant the reasonableness of predicted ozone enhancement (e.g., in Fig. 9) on the global scale due to the lack of wide-range calibrations.

Yes – this is the conclusion we reach as well: global models are not the best tools for estimating wildfire-ozone production in the near-field given that near-field plume chemistry is not well-represented at coarse resolution (L579):

> "In the near-field, we show that GEOS-Chem cannot capture concentrated fire emissions, and that the model cannot capture the NOx-suppressed and photochemically-suppressed sub-grid conditions."

After exposing the limitations of the model under this context, we use it to estimate the global wildfire-ozone burden for a single year, 2020. We are clear that this represents the modeled burden within GEOS-Chem. This is highlighted in the first sentence of Section 3.3.2 (L541):

> "In Figure 9, we quantify the overall impact that fires have in governing the tropospheric ozone burden in the GEOS-Chem model."

This estimate provides some useful context on the contribution of fires to ozone in global models and, in simultaneously revealing some of the shortcomings of the model in our work, we provide the reader with some insight into potential limitations of these estimates.

Third, the global attribution of 2.4% of the tropospheric ozone burden and 3.1% of surface ozone concentrations to fire emissions in 2020 may mask large regional and interannual variability. Fire activity is strongly influenced by El Niño–Southern Oscillation, land-use change, and climate variability. Using a single year could underestimate the range of fire impacts on global ozone. Moreover, uncertainties in fire emissions inventories, especially in Africa and South America, propagate directly into modeled ozone impacts but are not explicitly discussed.

We thank the reviewer for their feedback and agree that this is an important point. We have added text to Section 3.3.2:

> "It is worth noting that fire emissions exhibit interannual variability (Cooper et al., 2024) and that fire emissions inventories struggle with regionally-dependent uncertainties (Wiedinmyer et al., 2023). The reported results describe GEOS-Chem's representation of fire impacts on global ozone in a single year (2020), subject to the uncertainties in fire emission inventories."

SPECIFIC

Line 94: Previous studies have shown that "a smoke plume begins in a NOx-saturated regime before transitioning to a NOx-limited regime within a few hours." Since this is also a key conclusion of the present study, the authors should explicitly clarify what advances their work provides beyond prior findings.

Thank you for your comment. Previous studies have modeled this behavior or hypothesized its existence from analysis of limited data, but we believe that our study presents the first robust observational evidence of the regime transition using the unique, broad dataset we compile. This is mentioned in L320, beginning:

> "This confirms the theoretical predictions in previous studies…"

Section 2.4: What are the input factors used in the box model? How does the model account for potential impacts of meteorological variability or plume height on ozone formation?

The input factors are shown in Fig 7b. As mentioned in Section 3.2.1, "We use our box model to test the sensitivity of ozone production against specific factors including (1) the speciation and reactivity of the emitted *VOC* population, (2) the range of wildfire emissions observed across fires that vary in size, fuel, and burning conditions, (3) the emission of radical precursors, (4) the evolving availability of NO_x, and (5) aerosol effects." The corresponding factors are:

- (1) VOC/CO
- (2) CO, CH2O/CO
- (3) HONO/CO
- (4) NOx/CO
- (5) photolysis suppression (I), HO2 uptake onto aerosol (ASA)

The goal of our box modeling work is to compare the relative importance of different chemical factors in governing ozone production, regardless of physical parameters of the plume or background atmosphere, such as temperature or solar zenith angle, as discussed above. The ensemble of box model simulations discussed in this study is run at constant temperature (288.15K). While it is true that changing temperature governs the total ozone production, it does not change the relative importance of the different factors discussed. To illustrate this, we re-run our set of simulations at a lower temperature that roughly corresponds to a plume injection height of 4km (248 K). The results of this sensitivity test is shown in Figure S10. While the magnitude of PO3 changes slightly with the temperature change, the relative spread associated with each factor remains similar in both cases. See line 230:

> "Default photolysis rates are kept constant and calculated using the simplified MCM parameterization assuming a solar zenith angle of 0°, simulating clear-sky photolysis with the sun directly overhead. Temperature is held constant at 288.15 K. Our findings are robust to variations in temperature and solar zenith angle, as shown in Fig. S10."

Line 263: Why was a 10% variation chosen? For example, why not 50%?

The goal of the perturbation sensitivity study is to avoid “nonlinearities associated with the ozone chemical system, such as changing the oxidative potential of the global atmosphere” (L265), while also varying emissions enough to produce a noticeable impact (larger than numerical noise). One example of a past study that used the perturbation approach is the Hemispheric Transport of Air Pollution (HTAP) modeling experiments, which were focused on O_3 and use a 20% perturbation (Huang et al., 2017). Another is Carter et al., 2022, which uses a 5% fire emissions perturbation. We opt for a 10% perturbation, and we hypothesize that if we used a perturbation of 50%, we would likely see nonlinear chemistry effects emerge, which would complicate the fire source attribution.

Lines 292-299: These equations and related descriptions should be moved to Method.

Thank you for your suggestion. The text has been moved to Section 2.6 in the Materials and Methods.

Line 335: What is the sample size for each group?

The exact size of each regime has been added in Section 2.2 (L180).

> “The majority of our analysis focuses on the fire regime, which contains 4042 points, or 39% of the polluted subset (Fig. S4). The anthropogenic regime contains 3242 points (31%), and the mixed regime contains 1552 points (15%).”

Lines 347-348: “In general, *NOy* concentrations are largest in mixed air masses, followed by fire, then anthropogenic” It seems that the NOy concentrations are lower in mixed than fire as shown in Fig. 5b?

Thank you for catching this. We have changed the text to reflect the correct NOy regime comparison.

Lines 476-477: To what extent can increasing resolution and/or emissions be expected to improve the model’s predictability for near-field plumes?

It is generally difficult to separate emissions from resolution when evaluating a chemical transport model. As our work is more evaluative than mechanistic (see response to previous comment), answering this question is outside the scope of this study.

As cited in our manuscript, Wang et al. 2021 compares a series of large eddy simulations coupled to a chemistry model run at different horizontal resolutions (4km, 1km, 0.1km) to address the resolution question. Using the Williams Flats Fire as a case study, they find that (as stated on line 610):

> “…while a resolution of 1km was sufficient for capturing downwind chemistry, a model with 4km resolution failed at representing chemical regime shifts and thus incorrectly estimated O_3 formation. .”

As we further state in line 611: "With current computational constraints, GEOS-Chem and other global chemical transport models cannot operate at such fine resolutions."

So while extremely fine resolution does generally improve simulation performance, global models face a challenge in representing near-field plumes. Sub-grid parameterizations, such as the GEOS-Chem solution for ship plumes (Vinken et al., 2011), provide a potential workaround

We demonstrate an underestimate in emissions of both CO and NOx within GEOS-Chem. If these emissions were to be scaled to the physical values, it is unlikely that the bias discussed in Wang et al. 2021 would be corrected, since the emissions would still be diluted within a grid cell. However, the precise improvement offered by having accurate time-varying emissions remains unknown.

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